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Nanoindentation studies on waveguides inscribed in chalcogenide glasses using ultrafast laser

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Abstract: Optical straight waveguides are inscribed in GeGaS and GeGaSSb glasses using a high repetition-rate sub-picosecond laser. The mechanical properties of the glasses in the inscribed regions, which have undergone photo induced changes, have been evaluated by using the nano-indentation technique. Results show that the hardness and elastic modulus of the photo-modified glasses are significantly lower as compared to the other locations in the waveguide, which tend to be similar to those of the unexposed areas. The observed mechanical effects are found to correlate well with the optical properties of the waveguides. Further, based on the results, the minimum threshold values of hardness and elastic modulus for the particular propagation mode of the waveguide (single or multi), has been established.

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1. Introduction

Chalcogenide glasses (ChGs) are an important class of amorphous semiconductors due to their unique optical properties especially their high third order optical non-linearity $\chi^{(3)}$ compared to other glasses ($\sim 10^3$ times higher than that of a-SiO₂) [1], good transmittance in the infrared region (sulphides $\sim 11\ \mu\text{m}$, selenides $\sim 15\ \mu\text{m}$ and tellurides $\sim 20\ \mu\text{m}$) [2] and low phonon energy [3]. They are also good host materials for rare earth ions. ChGs also exhibit a variety of photo-induced phenomena not normally observed in crystalline chalcogenides [4]. Due to these interesting properties, ChGs are widely used in electronic, photonic and optoelectronic industries in several applications such as IR optical devices [5], amplifiers and lasers [6,7], phase change memory devices [8] and optical waveguides [9,10].

Recently, concerted efforts have been made to fabricate three dimensional (3D) photonic integrated devices in ChGs using the ultrafast laser writing technique [9,11,12]. The direct laser writing (DLW) technique allows to fabricate integrated optical elements such as gratings or optical waveguides at the surface or deep inside ChGs, without the need for complex lithography and clean room facilities [13]. It is based on the principle that within the focal volume of an ultra-short laser pulse, the structure of the glass can get modified due to multi-photon and/or avalanche nonlinear ionization, so that the modified region will have a smaller volume than that of the focal zone if the fluence is kept close to threshold. It is also known that the accumulation of thermal energy that occurs during laser writing produces a heat-modified volume which can be much larger than that of the photo-excited region. In

general, the final degree of modification of the glassy matrix, as well as its volume, will depend on the interplay of many different factors, such as the laser pulse repetition rate, the effective exposure time, the resultant heat accumulation, the heat diffusion from the active volume [14], the glass properties, the temperature-dependence function of the glass viscosity [15], the free carrier accumulation [16], etc.

Generally, photo induced modifications are associated with changes in density and refractive index [17], mass transfer [18], the fluidity of the glasses [19], etc. Understanding the precise mechanisms which drive the photo-induced changes in a chalcogenide glasses is essential for the development of active and passive photonics devices for wavelengths from the visible to the mid-infrared [20,21]. In the present work, we employ the nano-indentation technique, which allows measurement of the mechanical properties of small volumes of materials, to characterize optical straight waveguides inscribed in GeGaS and GeGaSSb glasses using femto-second laser pulses. Importantly, we show that it can also be utilized to identify the right combination of process parameters which would produce good waveguides.

2. Experiments

2.1 Materials synthesis

Bulk GeGaS and GeGaSSb glasses have been prepared by vacuum-sealed melt quenching technique. Appropriate quantities of high purity (99.999%) constituent elements are sealed in flattened quartz ampoules under a vacuum of 10^{-5} Torr. The sealed ampoules are subsequently heated in a rocking furnace to a temperature above the melting temperature of the constituents at a heating rate of about 100 °C/h. The ampoules containing the melt are rotated continuously at 10 rpm for about 12 h, to ensure the homogeneity of the melt. The ampoules are subsequently cooled down in air to obtain optical quality bulk glasses.

2.2 Laser inscription of waveguides

The waveguides have been written in a GeGaS glass sample of 5 mm length using an amplified Yb-doped fiber femtosecond laser (IMRA μ jewel D400). During inscription, the pulse repetition rate and polarization are set at 100 kHz and circular respectively. The pulse duration is 440 fs and the central wavelength of the laser radiation is 1047 nm. Different straight waveguides have been written 100 μ m below the surface of the glass using a microscope objective with a numerical aperture of 0.9 NA. Different laser pulse energies from 276 to 62 mW, and translation speeds of 1, 2, 3 and 4 mm/s, have been utilized. The experimental details of the laser inscription of waveguides are described elsewhere [22].

After fabrication, the sample facets are polished to optical quality to reveal the waveguide cross-sections. All the photo induced tracks appear to clearly guide light both in the visible and near-infrared, characterized by an increased refractive-index, with a morphology which suggests the thermal accumulation phenomena as one of the main driving modification mechanisms.

2.3 Nanoindentation studies

Nanoindentation studies are performed on the samples using the Triboindenter (Hysitron, Minneapolis, USA) with *in situ* imaging capability. The machine continuously monitors the load P and the depth of penetration h of the indenter with resolutions of ~ 1 nN and ~ 0.2 nm, respectively. A Berkovich tip diamond indenter with a tip radius of ~ 100 nm is used for indentation. A peak load P_{max} of 8 mN with loading and unloading rates of 0.8 mN/s and a hold time (at P_{max}) of 10 s is employed. Post-indentation images of the impressions are captured immediately. A minimum of eight indentations have been performed in each case and the average of them is reported. The P - h curves are analyzed using the Oliver-Pharr method [23] to extract the elastic modulus, E , and hardness, H , of the glasses. The detailed methodology is given in [24].

2.4 Optical characterization

The waveguide mode near-field image at 1550 nm wavelength and the waveguide core structure (under visible light optical microscope examination) are shown in Figs. 1(a) and 1(b) respectively. Further details about the waveguides have been provided elsewhere [11].

In Fig. 1(a), the tear drop waveguide structure seen is due to the geometry used for fabricating the waveguide. The transverse geometry is advantageous as in the longitudinal inscription, the length of the waveguide is limited to the stage travel range. However, the transverse geometry suffers from the disadvantage of asymmetry in the shape of the waveguide (tear-drop); the asymmetry occurs because of the fact that the focus diameter of the laser beam ($2\omega_0$ few μm , where ω_0 is the radius of the focus diameter) is much smaller than the confocal parameter ($b = 2\pi\omega_0^2/\lambda$) [25], where, λ is the wavelength of the laser. In this context, it is interesting to note that there are earlier studies which discuss the asymmetry and the parameters that control the spatial formation of the direct laser written waveguide [26,27].

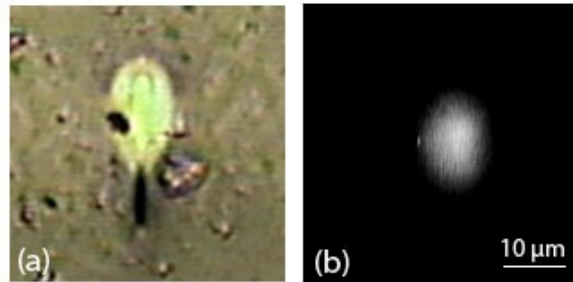


Fig. 1. (a) An optical micrograph of the single mode waveguide structure with 50X magnification under white light illumination and (b) its near field image at 1550 nm wavelength.

3. Results and discussion

Measured values of E and H indicate to three distinct regions within the inscribed waveguide (Fig. 2(a)) and outside the waveguide; region I which corresponds to the whole waveguide structure and region II corresponding to the top of the tear-drop (identified as position “0” in Fig. 2(a)). The region III pertains to remaining region, unexposed to light i.e., outside the waveguide. The representative load, P , vs. displacement, h , curves obtained from the three regions are shown in Fig. 2(b). The measured values of E and H at different positions in the waveguide are shown in Fig. 2(c). It is observed that both E and H are similar at different places of the waveguide and the region III, except at position “0” wherein they are nearly half of the bulk values. While the average values of E and H at different places of the waveguide are 30 and 3 GPa, respectively, they are 15 and 0.75 GPa respectively at position “0”. Interestingly, not much difference is observed in the properties in the laser exposed and unexposed regions. Both the regions exhibit a smooth P - h behaviour with significant levels of residual depths upon complete unloading. Images of the indentation impressions show neither corner cracking nor pile-up of the material against the indenter.

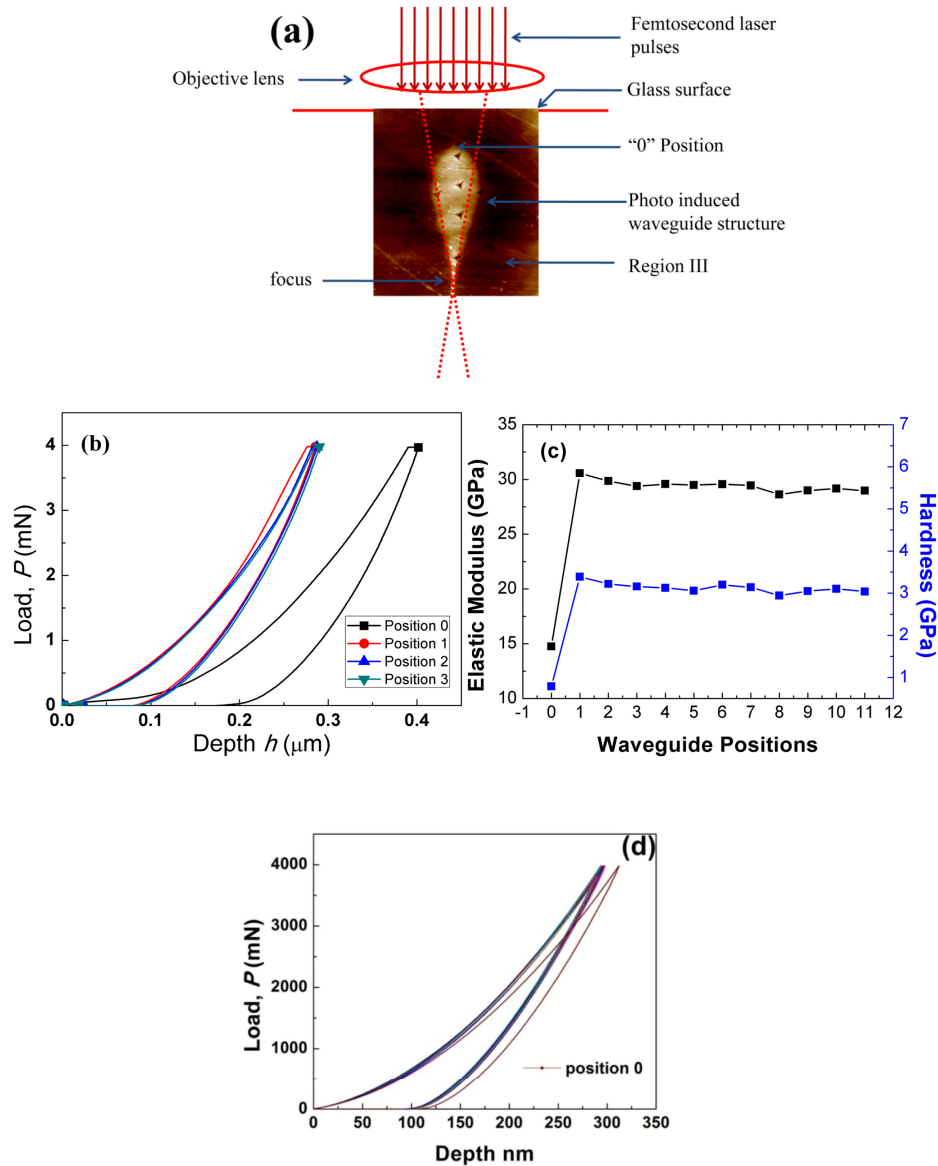


Fig. 2. (a) Geometry of the waveguide. (b) Representative load, P , vs. displacement, h , curves at different locations of the waveguide on GeGaS glass (c) Variation of E and H with the positions in the waveguide and (d) Representative load, P , vs. displacement, h , curves at different locations of the waveguide on GeGaSSb glass.

The extent of the laser induced structural modification is gauged by the difference between the E and H values measured at the position "0" and the average E value obtained in other regions (both exposed and unexposed). This difference is denoted as ΔE and ΔH respectively. Based on ΔE and ΔH , it can be suggested that the material at 0th position is severely modified by the laser and the same was reflected as ΔE and ΔH .

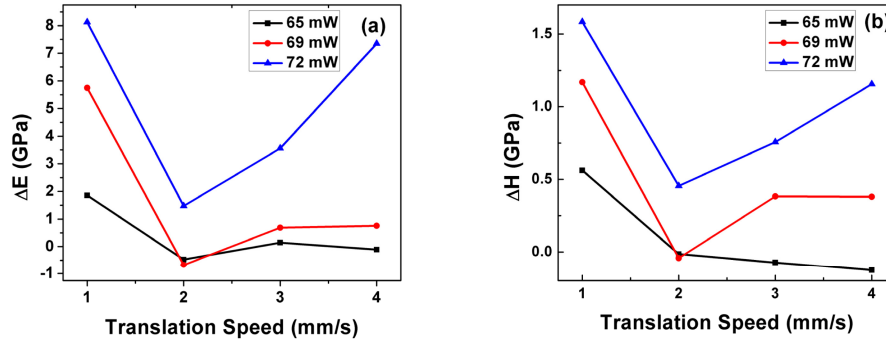


Fig. 3. Variation of (a) ΔE and (b) ΔH with varying the laser translation speeds at position zero.

Variations in ΔE and ΔH with the translation speed of the laser and at different laser energy levels are plotted in Figs. 3(a) and 3(b). From these plots, it is seen that in general higher energy leads to higher values of ΔE and ΔH ; also, low translation speeds (< 2 mm/s) results in a significant drop in both E and H at position “0”, with the increase in translation speeds. However, above 2 mm/s, an increase in ΔE and ΔH is observed, especially at higher energy levels employed (72 and 69 mW). However, at 65 mW energy, there is no significant change in ΔE and ΔH with translation speeds above 2 mm/s. Thus, an energy of 65 mW with translation speeds between 2 and 4 mm/s appear to optimal from the viewpoint of obtaining the ‘uniform’ waveguide.

It is also interesting to examine the correlation between the measured variations in mechanical properties and the optical characteristics of the waveguides. It is found that when the values ΔE and ΔH are below 1.85 and 0.56 GPa, respectively, the waveguides support a “single mode” at 1550 nm. Above these values, the waveguides support multi-modes. These results are in conformity with the near field images shown in Fig. 1(b).

In order to examine the repeatability and universality of the present observations in other chalcogenide glasses, nanoindentation studies have been performed on waveguides written on GeGaSSb glass under the similar experimental conditions. The GeGaSSb waveguides are written using DLW technique and with the repetition rate at 500 kHz and the energies are varied from 183 mW to 66 mW. The results observed indicate that the effects seen in GeGaS glass waveguide are also seen in GeGaSSb glass waveguide which is shown in Fig. 2(d).

Earlier studies on the mechanical properties of chalcogenide glasses reveal that H and E change during under band-gap illumination [28,29], which has been attributed to thermal annealing. However, the present results reveal that there is the difference in mechanical properties of the material at light exposed and unexposed regions is insignificant, except at position “0”. In glasses, reduction in E and H values indicate significant density reductions. Thus, the lower mechanical properties at position “0” imply relatively less denser (or open) structure. Further, the present study clearly indicates that certain combinations of translation speed and input energy will result in such local structural effects in the glass. The possible reasons for this are discussed below.

It is likely that during the waveguide writing process using femto-second laser pulses, the material melts locally around the focal zone due to the high temperature created by the laser pulses. Once the laser focus is shifted to the subsequent point for creating the waveguide, the molten material re-solidifies. This process is similar to melt quenching technique which is used for synthesizing the different glassy materials including chalcogenides; here, the quenching takes place immediately after exposure of light. Consequently, the observed mechanical properties at the focal zone of the exposed region and un-exposed regions are nearly the same.

However, when position “0” is approached, the probability of multi photon absorption decreases and also the beam size increases in comparison with the focal zone. For comparison, at the focal point, the beam diameter is $\sim 0.5\ \mu\text{m}$ and at the “0” position the beam diameter is 3 mm. At 100 kHz repetition rate in 1 mm/s translation speed, at the “0” position, thermal energy stays ~ 1000 times more than at focuses because of the laser beam diameter and the overlapping of the beam. As a consequence, intense heating is not likely and the thermal energy stays for a longer duration in this region due to the larger beam size. Consequently, the thermal process which takes place at position “0” is closer to annealing than quenching; the structural reorganization associated with thermal annealing, leads to the observed change in mechanical properties at position “0”.

It is also interesting to note here that, both the effects of annealing and quenching have been observed for the first time in the present study, during a single exposure. In this work, the spatial extent of the “0” position has also been investigated, by choosing the indent points with 1 μm separation which is limited due to the residual impression of the indenter. It is found that the extent of the “0” position is $\sim 3\ \mu\text{m}$ for the 20 μm diameter waveguide.

4. Summary

In summary, we have fabricated waveguides on GeGaS and GeGaSSb glass using Yb-doped fiber femtosecond laser. The waveguides are written with different laser energies and translation speeds. The mechanical properties at different places of the waveguides have been measured using nanoindentation. It is found that the change in the mechanical properties such as E and H are position dependant in the waveguides. Based on these results, we have proposed the cut-off values for ΔE and ΔH , for supporting single modes at 1550 nm. From the mechanical properties, we could characterize the waveguides to find suitable single mode waveguide at 1550 nm.

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